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Abstract: We describe the experimental observation of ozone formation inside an air-filled hollow-core fiber driven by ultra-short pulses and its effect on the emitted UV resonant dispersive wave.

1. Introduction

Gas-filled hollow-core photonic crystal fibers (HC-PCF) have been used extensively in the last decade to develop new light sources and to study the nonlinear interaction between laser light and gases [1, 2]. The long interaction length, dispersion tunability, and broadband transmission window allowed the use of HC-PCF in studying ultrafast nonlinear phenomena such as supercontinuum generation, resonant dispersive wave (RDW) emission, light-plasma interaction, and soliton self-compression. In this work, we show experimental evidence of the formation of ozone (O_3) inside air-filled HC-PCF, as a result of Chapman reactions [3]. Ozone formation is triggered by ultrafast oxygen photo-dissociation ($O_2 \rightarrow O + O$), followed by recombination with molecular oxygen ($O_2 + O \rightarrow O_3$). Ozone exhibits a broad ultraviolet absorption band around 255 nm, which strongly affects both the propagation loss and the dispersion in the PCF. We observed periodic evolution in the energy of a UV RDW generated in this spectral region on the timescale of tens of seconds. Moreover, the RDW showed a red-shift in its central wavelength. We numerically investigate these phenomena by solving the unidirectional pulse propagation equation in combination with the Chapman reaction equations. Our simulations show how the RDW central wavelength changes as the amount of ozone in the fiber increases.

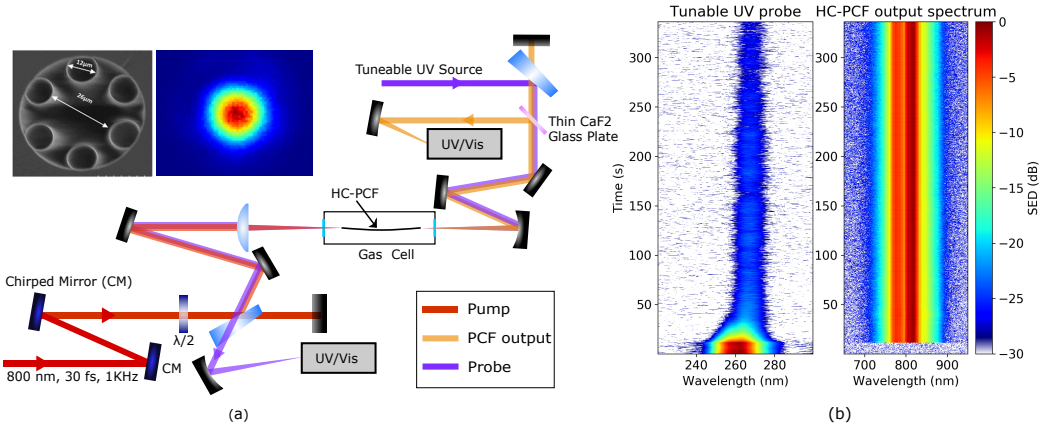

Fig. 1: (a) Experimental setup. The insets show a scanning electron micrograph of the fiber cross-section and a near-field image of the fiber output mode. (b) Results of ozone formation investigation for one bar of air and a pump pulse energy of 1.5 μJ. The left plot shows the UV probe (centered around 260 nm, close to ozone absorption peak at 255 nm) transmitted through the PCF and the right plot shows the PCF output spectrum. For the first 15 seconds, the input beam was blocked (so no ozone was formed inside the fiber). After this point, the pulses were launched into the fiber, causing ozone to be formed and the probe to be absorbed.

2. Experimental Setup and Results

Fig. 1a shows the experimental setup. A 30 fs pump pulse centered at 800 nm is launched into a 23 cm long anti-resonant HC-PCF with 13 μm core radius filled with air up to a pressure of 12 bar. The pulse undergoes soliton self-compression since the dispersion of the fiber is tuned to be anomalous for the pump wavelength. As the pulse compresses, its peak intensity increases up to 10^{14} W/cm^2 inside the fiber. At such high intensity, oxygen molecules can dissociate, forming two oxygen atoms [4]. Following the Chapman cycle, these oxygen atoms can then interact with oxygen molecules to form ozone molecules. We have investigated the signature of ozone formation experimentally via absorption spectroscopy using a tuneable UV probe. With the pump pulse unblocked, the probe pulse was strongly absorbed around 255 nm as shown in Fig. 1b, matching the ozone absorption cross-section. In this experiment, we used 1.5 μJ pump pulses and filled the fiber with one bar of air. At these parameters, RDW emission does not occur, so oxygen molecules dissociate only due to the strong field at 800 nm.

In a second experiment, we filled the fiber with 6 bar of air and used 1.8 μJ pump pulses. At this pressure and energy, the pulse undergoes soliton fission and a UV RDW is generated. The RDW is centered at around 280 nm initially. As time progresses, more oxygen molecules dissociate, hence more ozone can form, which leads to a change in the dispersion and loss profile seen by the pulse. The central wavelength of the RDW shifts toward longer wavelengths. Fig. 2a shows how the experimental output spectrum changes with time. During the first few seconds, the RDW central wavelength shifts towards longer wavelengths as shown on the inset of Fig. 2a. Then, the RDW appears and disappears periodically in time at the original central wavelength (280 nm). The change in the RDW central wavelength is due to the change in the dispersion caused by the ozone. We have numerically investigated this phenomenon by solving the unidirectional pulse propagation equation for different uniform concentrations of ozone. We used the Kramers-Kronig relations to calculate the dispersion profile of ozone from its absorption cross-section for our numerical simulations. Fig. 2b shows simulations of how the RDW wavelength changes for different percentages of ozone.

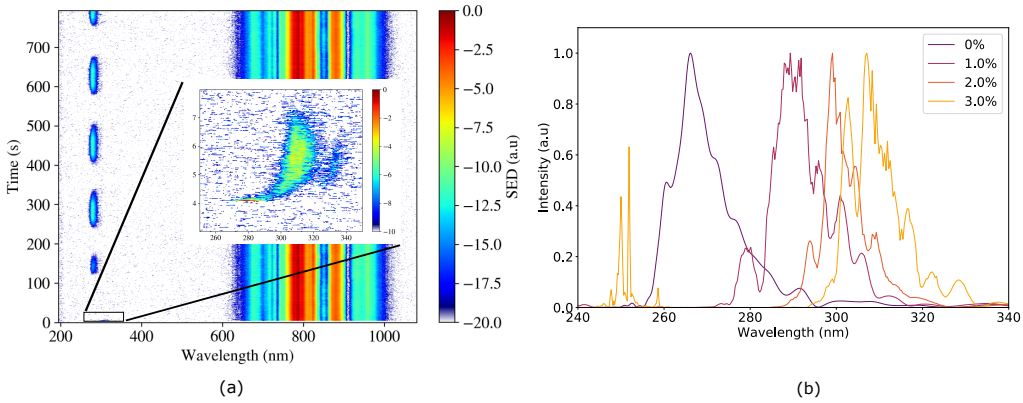

Fig. 2: (a) Experimental output spectra with time for 6 bar air and 1.8 μJ pump pulse energy. The inset shows the spectrum of the RDW in the first few seconds. (b) Numerically simulated output spectrum of the RDW for different uniform ozone concentrations inside the fiber without including the ozone loss. The total pressure of air and ozone is kept constant at 6 bar.

Since the pulse duration and peak intensity is not constant during the propagation inside the fiber, due to the soliton dynamics, the concentration of ozone will also not be constant along the fiber. As a result, the ozone may diffuse along the fiber, potentially causing the periodic pattern of the RDW shown in Fig 2a. Further investigation of these dynamics is required to fully explain the periodic pattern of the RDW in time.

References

1. Russell, P. St.J., P. Hölzer, W. Chang, A. Abdolvand, and J. C. Travers. “Hollow-Core Photonic Crystal Fibres for Gas-Based Nonlinear Optics“. Nature Photonics 8, 278–286 (2014).
2. Travers, John C. *et al*. “Ultrafast Nonlinear Optics in Gas-Filled Hollow-Core Photonic Crystal Fibers [Invited]“. J. Opt. Soc. Am. B 28, A11–A26 (2011)
3. Chapman. “On ozone and atomic oxygen in the upper atmosphere“. The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science 10, 369–383 (1930).
4. Song, D. *et al*. Neutral Dissociation of Superexcited Oxygen Molecules in Intense Laser Fields. J. Phys. Chem. A 114, 3087–3095 (2010).